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THE THEORY OF THE PROPAGATION OF LIGHT IN POLYCRYSTALLINE MEDIA

By Sir C. V. Raman and K. S. Viswanathan
(*Memoir No. 66 of the Raman Research Institute, Bangalore*)

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1. Introduction

Many common minerals occur in nature as polycrystalline aggregates. For example, quartz, calcite and gypsum appear in massive form respectively as quartzite, marble and alabaster, and on examining thin sections of these materials under the polarisation microscope, it is found that they consist of great numbers of crystallites variously orientated and firmly adherent to each other so as to form a coherent solid. The size, shape and manner of orientation of the crystallites may differ enormously in individual cases. Some minerals are indeed cryptocrystalline, in other words, the particles are so small that they cannot be identified by the usual polariscopic methods and require the aid of X-ray analysis to enable their true nature to be determined.

The foregoing is by way of stressing the importance alike to the mineralogist and to the physicist of a study of the physical properties of polycrystalline aggregates. It is obvious that the optical properties of the single crystal and especially its birefringence and pleochroism (if any) would play a dominant role in determining the optical characters of the polycrystalline aggregate. Considering the matter from the standpoint of geometrical optics, it is evident that when light enters a polycrystalline aggregate, it would suffer reflection at the intercrystalline boundaries. The stronger the birefringence, the greater would be the coefficient of reflection at these boundaries and hence the more quickly would the incident light be returned back towards the source. The brilliant whiteness of pure marble is thus a recognisable consequence of the strong birefringence of calcite. On the other hand, if the birefringence be feeble as in the case of quartz and gypsum, the incident light would penetrate far more deeply into the aggregate. Ultimately, all the light would necessarily be turned back provided that a sufficient thickness of the material be available and that no absorption intervenes. If the thickness of the material be insufficient, a part of the light would diffuse out—a phenomenon readily observed with various materials. Light

so emerging would be depolarised even if the light incident on the plate were fully polarised in the first instance.

Various considerations indicate that a purely geometric theory is inadequate to cover all the optical phenomena actually exhibited by polycrystalline media. It will suffice to mention here the question of the influence of the size of the crystallites. Geometric considerations would suggest that the smaller the crystallites, the more numerous would be the reflections and refractions at the intercrystalline boundaries, and hence the more rapidly would the incident light be diffused and extinguished in its passage through the medium. Experience however suggests that the contrary may actually be the case, and that the more fine-grained the material is, the more deeply would the light penetrate into it. Various minerals, *e.g.*, alabaster and jade, which exhibit marked translucency, are usually fine-grained; the finer the grain, the more deeply does light penetrate into them. This suggests that the optical problems presented by polycrystalline aggregates require to be considered from the standpoint of the wave-theory of light. That indeed is the object of the present paper.

2. A SIMPLIFIED MODEL

To obtain some results of physical interest and also with a view to simplify the mathematics, we shall here restrict ourselves to the case of a feebly birefringent material and consider the case in which light is incident normally on a plate with parallel faces; this is assumed to be sufficiently thick to include a great many individual crystallites but not so thick that the incident light is completely extinguished before it can emerge at the rear face. We may disregard the geometric course of the individual rays of light and view the matter purely from the wave-theoretical standpoint. Owing to the varying orientation of the individual crystallites, the waves of light entering the plate would be retarded to different extents in passing through them. To enable the resulting total retardation to be evaluated, we use a simplified model and assume the plate to be an assembly of a great number of small cubical blocks each having a common edge-length Δ and completely filling up the available space. Each block is assumed to be a single crystallite, and the three edges of each cube to be parallel to the three optic directions for which the refractive indices are μ_1, μ_2 and μ_3 respectively. To introduce the idea of varying orientation and to take account of its influence on the propagation of light through the material, we assume the incident light-beam to be plane-polarised with its vibration direction parallel to one set of edges of the cubical blocks; on the other hand, the operative refractive index of any one block may be either μ_1 or μ_2 or μ_3, the respective

probabilities for these being p_1, p_2, p_3. The case where the three probabilities are equal would correspond to a random orientation of the crystallites in the present restricted sense of that term. More generally, by giving appropriate values to p_1, p_2, p_3 such that their sum remains equal to unity, we obtain a representation of a polycrystalline aggregate with any desired measure of preferred orientation along the particular direction under consideration. If, for example, we put $p_1 = 1$ while p_2 and p_3 are zero, it would mean that all the particles of the aggregate have a common refractive index for the particular direction of vibration, though the indices may be different in the perpendicular direction.

On the assumptions stated, the incident plane-polarised disturbance would remain plane-polarised in its passage through the plate, though subject to phase retardations of varying extents. The situation would no doubt be different for any actual polycrystalline material, since the incident plane-vibration would be transformed to an elliptic vibration and the parameters describing the ellipticity would alter as the disturbance passes from crystallite to crystallite. While it would no doubt be possible to deal mathematically with this general case, a very considerable simplification is effected by our present assumptions, and as we shall see, the usefulness of the results obtained is not affected thereby.

A further question needing consideration is the effect of the reflections which would occur at the boundary between every two successive blocks. This would obviously diminish the amplitude of the transmitted disturbance. As a first approximation, we may assume such diminution to be the same over all the individual elementary areas Δ^2 on the rear surface of the plate and represent it by a numerical factor of appropriate magnitude. In other words, we ignore the variation of *amplitude* over the different elementary areas Δ^2 on the rear face of the plate and consider only the variations of *phase*. The latter are in reality of much greater importance for the determination of the final observable result.

3. Mathematical Formulation

Let us suppose that the wave-train before entry into the plate is represented by

$$y = e^{\frac{2\pi i}{\lambda}(vt - Z)} \tag{1}$$

and that there are n cells along the direction of the thickness of the plate. We shall first consider a typical case in which the wave has passed through k_1 cells of refractive index μ_1, k_2 cells of refractive index μ_2 and k_3 cells of

refractive index μ_3 before emerging from the plate. The numbers k_1, k_2 and k_3 can all vary from zero to n subject to the relation

$$k_1 + k_2 + k_3 = n \tag{2}$$

The optical path retardation of the emergent wave would then be equal to $(k_1\mu_1 + k_2\mu_2 + k_3\mu_3)\,\Delta$.

Now the number of ways in which k_1, k_2 and k_3 cells can be orientated along a row of n cells so as to have refractive indices μ_1, μ_2 and μ_3 is obviously

$$\frac{n!}{k_1!\,k_2!\,k_3!}$$

and the probability of occurrence of each one of these cases is $p_1^{k_1}p_2^{k_2}p_3^{k_3}$. Hence the proportion of the total area of the rear surface of the plate from which a wave represented by

$$e^{\frac{2\pi i}{\lambda}(ct - Z - \overline{k_1\mu_1 + k_2\mu_2 + k_3\mu_3}\,\Delta)} \tag{3}$$

emerges is equal to

$$\frac{n!}{k_1!\,k_2!\,k_3!}\;p_1^{k_1}p_2^{k_2}p_3^{k_3} \tag{4}$$

The emergent wave-train can now be obtained by summation of waves of the type (3) with their appropriate amplitudes and phases for all possible integral values of k_1, k_2 and k_3 satisfying the relation (2). We therefore have for the emergent wave

$$y = \mathrm{P}\sum_{k_1+k_2+k_3=n} \frac{n!}{k_1!\,k_2!\,k_3!}\;p_1^{k_1}p_2^{k_2}p_3^{k_3}\;e^{\frac{2\pi i}{\lambda}(ct - Z - \overline{k_1\mu_1 + k_2\mu_2 + k_3\mu_3}\,\Delta)} \tag{5}$$

where P is a factor which is introduced to take into account the loss in intensity of the light due to reflections at the intercrystalline boundaries.

In view of the multinomial theorem, equation (5) may be rewritten as

$$y = \mathrm{P}\,e^{\frac{2\pi i}{\lambda}(ct - Z)}\left(p_1 e^{\frac{-2\pi i\mu_1\Delta}{\lambda}} + p_2\,e^{\frac{-2\pi i\mu_2\Delta}{\lambda}} + p_3 e^{\frac{-2\pi i\mu_3\Delta}{\lambda}}\right)^n \tag{6}$$

The average refractive index of the medium is clearly

$$\mu = (p_1\mu_1 + p_2\mu_2 + p_3\mu_3) \tag{7}$$

If therefore we set $\nu_1 = (\mu_2 - \mu_3)$; $\nu_2 = (\mu_3 - \mu_1)$; and $\nu_3 = (\mu_1 - \mu_2)$, we can then express μ_1, μ_2 and μ_3 as

$$\mu_1 = \mu + (p_2\nu_3 - p_3\nu_2)$$
$$\mu_2 = \mu + (p_3\nu_1 - p_1\nu_3) \tag{8}$$
$$\mu_3 = \mu + (p_1\nu_2 - p_2\nu_1)$$

Further, the thickness of the plate is given by $d = n\Delta$. Hence substituting the relations (8) in (6) and expanding the exponential terms in power series of their arguments, one obtains

$$y = \mathrm{P}\, e^{\frac{2\pi i}{\lambda}(ct - Z - \mu d)} \times \left\{ 1 - \frac{2\pi^2\Delta^2}{\lambda^2} \Sigma p_1 (p_2\nu_3 - p_3\nu_2)^2 \right\}^n \tag{9}$$

As the birefringence is assumed to be small, we have ignored terms of third and higher powers of $(\mu_1 - \mu_2)$, $(\mu_2 - \mu_3)$ and $(\mu_3 - \mu_1)$ in (9). Also by means of a small simplification, it can be verified that

$$\Sigma p_1 (p_2\nu_3 - p_3\nu_2)^2 = \Sigma p_2 p_3 \nu_1^{\,2}$$

Hence, we can rewrite (9) as

$$y = \mathrm{P}\, e^{\frac{2\pi i}{\lambda}(ct - Z - \mu d)} \left\{ 1 - \frac{1}{n} \times \frac{2\pi^2 \Delta d}{\lambda^2} \Sigma p_2 p_3 \nu_1^{\,2} \right\}^n$$

$$= \mathrm{PR}\, e^{\frac{2\pi i}{\lambda}(ct - Z - \mu d)}$$

where

$$\mathrm{R} = e^{\frac{-2\pi^2 \Delta d}{\lambda^2} \Sigma p_2 p_3 \nu_1^2} \quad \text{as } n \text{ is large.}$$

The ratio of the intensity of the transmitted light to that of the incident radiation is therefore given by

$$\frac{\mathrm{I}}{\mathrm{I}_0} = \mathrm{P}^2\mathrm{R}^2 = \mathrm{P}^2\, e^{\frac{-4\pi^2\Delta d}{\lambda^2} \Sigma p_2 p_3 (\mu_2 - \mu_3)^2} \tag{10}$$

If the three optic axes of any cube have the same probability of being orientated in the direction of the incident light, then $p_1 = p_2 = p_3 = \frac{1}{3}$ and (10) reduces to

$$\frac{\mathrm{I}}{\mathrm{I}_0} = \mathrm{P}^2\, e^{\frac{-8\pi^2\Delta d}{9\lambda^2} (\Sigma \mu_2^2 - \Sigma \mu_2 \mu_3)} \tag{11}$$

4. Significance of the Results

The physical meaning of the result stated in (10) is that the plane-polarised waves incident on the front of the plate emerge from the rear face

of the plate also as a plane-polarised vibration but with an attenuated amplitude determined by an exponential factor involving four variables, namely the size of the particles, the thickness of the plate, the wave-length of the light and a quantity which is a measure of the birefringence of the material, since it vanishes when the three indices μ_1, μ_2, μ_3 are all equal. The appearance of λ^2 in the denominator indicates that white light entering the plate would emerge enfeebled but with the longest waves predominant, in other words, much reddened in colour. Since the wave-length λ is a small quantity, the actual intensity of the emerging light would be negligible if both Δ and d are large. The individual crystallites have, in fact, to be quite small and the total thickness traversed should be moderate if any observable fraction of the light is to emerge as a coherent optical beam. We have already assumed the birefringence to be small and the need for such assumption is reinforced by our final result which indicates that unless the three indices μ_1, μ_2, μ_3 differ from each other by quantities which are small fractions of their absolute magnitudes, no light can emerge from the rear of the plate.

We may illustrate the foregoing remarks by the case of a plate of alabaster 1 mm. thick taking λ and Δ equal to 5896 A.U. and $1\,\mu$ respectively; the three indices for gypsum are $\mu_1 = 1 \cdot 520$, $\mu_2 = 1 \cdot 523$ and $\mu_3 = 1 \cdot 530$. The percentage of transmission then comes out as $13 \cdot 5\%$ but increases to 37% and 82% if $\Delta = 0 \cdot 5\,\mu$ and $0 \cdot 1\,\mu$ respectively. Thus, the formula indicates that a plate of alabaster approaches practically complete transparency as the crystallites of which it is composed approach colloidal dimensions.

The case of preferred orientations is also of interest in view of the known optical behaviour of chalcedony and of certain forms of gypsum. Taking now the general formula (10), if we put $p_1 = 1$ while p_2 and p_3 are both zero, the formula indicates that the transmission becomes complete. In other words, if the crystallites are so orientated that all of them have a common refractive index for the direction of vibration of the incident light, then we have a complete transmission of the incident light wave. But the position would be totally different for a perpendicular direction of vibration if the refractive indices for the latter direction differ from crystallite to crystallite. The general formula (10) would then show only a partial transmission depending upon the actual values of the probabilities and the refractive indices for that direction. Since the latter transmission would depend upon the thickness d, it would follow that if the light incident upon the plate be unpolarised, the state of polarisation of the emerging light would vary with the thickness of the plate. Formula (7) also indicates hat the effective refractive index of the medium would be different for the

two directions of vibration under consideration. Light which is plane-polarised in any arbitrary azimuth when incident on the plate, would emerge as elliptically polarised light, the parameters describing such ellipticity varying with the thickness of the plate—a phenomena readily capable of experimental verification with materials of the nature under consideration.

5. Some Further Remarks

The question naturally arises as to where the energy goes which disappears from the incident light according to (10). The answer to this is not far to seek. Since the reduction of amplitude is a consequence of the random variations of phase over the elementary areas of the rear face of the plate, the missing light would appear as diffracted radiation spread out in various directions surrounding the direction of the incident beam. The angular dimensions of the diffraction halo would obviously be comparable with the ratio between the wave-length λ of the light and the linear dimension Δ of the crystallites which we have assumed the material to be composed of. A diffusion halo of this type can indeed readily be observed on viewing a bright source of light through a thin plate of alabaster surrounding the sharply defined image of the source itself. According to the theory developed above, both the light emerging from the rear surface of the plate and the light appearing in the diffusion halo would be perfectly polarised if the incident light be itself plane-polarised. These results are consequences of the special assumption regarding the orientation of the crystallites which we have made. We may now ask ourselves whether they would continue to be true if the particles are orientated truly at random. The answer to this question is most readily ascertained by making a few actual observations with a plate of alabaster sufficiently thin to give a true transmission. It is then observed that the true transmission is completely polarised while the diffraction halo seen overlying it is imperfectly polarised. A more complete mathematical theory which takes account of the ellipticity resulting from the passage of light through an arbitrarily orientated crystal block would no doubt yield results in agreement with these facts of observation. It is clear, however, that the present theory suffices to indicate the state of polarisation of the transmitted light correctly and also its intensity, at least as regards the order of magnitude. But the theory fails to indicate the state of polarisation of the diffracted light accurately, since it ignores the ellipticity produced by the passage of light through a birefringent crystal in an arbitrary orientation; such ellipticity would obviously result in diverting some of the incident energy into the perpendicular component of vibration as diffracted radiation.

SUMMARY

A formula based on wave-theoretical considerations is deduced which gives the coefficient of extinction of plane-polarised light traversing a polycrystalline aggregate in terms of the wave-length of the light, the size of the particles and their birefringence. The general formula covers the case where the particles have preferred orientation expressible by three different probability numbers for three mutually perpendicular directions, and the special case of isotropic orientation is readily derivable therefrom. The significance of the results is discussed in relation to the facts of observation.

THE RANGE-ENERGY RELATION IN NUCLEAR EMULSIONS

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Received January 13, 1955

The use of large emulsion block detectors for the study of the masses and modes of decay of unstable particles has made it possible to measure the range of fairly energetic particles. In order to use such observations for good energy determinations an accurate knowledge of the relation between range and energy for different types of particles in nuclear emulsions upto fairly high energies is required. Energy values are needed for particles with masses equal to or greater than that of μ-mesons upto ranges of about 20 cm. Since range-energy relations for different singly charged particles can be directly transformed into one another provided their masses are known, it is sufficient to discuss the range-energy relation for protons. The required curve for protons should, therefore, extend to $20\times\frac{M_p}{M_\mu}\sim 180$ cm. or an approximate proton energy of 2 Bev.

Experimentally the range-energy relation in nuclear emulsions has been investigated by various workers[1-4] upto proton energies of 40 MeV and a corresponding residual range of about 6 mm. In restricted energy intervals these results can be represented by empirical relations of the type $E=kR^x$ where E is the energy, and R the range of the particle. Experimental data for protons with energies between 40 MeV and 2 BeV could be obtained with particles accelerated in large synchro cyclotrons, but at present, the only point on the range-energy curve measured in this energy interval is that obtained by Heinz[5] for 342·5 MeV protons. Therefore, in the absence of more extensive detailed measurements, it becomes necessary to construct a range-energy relation which will represent all existing experimental data and permit reasonable interpolation and extrapolation.

In using the available experimental results one has to bear in mind the dependence of the stopping power of emulsions on their moisture content. When reduced to equal moisture content it is found that the experimental range-energy data for emulsions are related in a simple manner to the corresponding theoretical values for Aluminium and Lead as calculated by

Smith[6] and published in the form of graphs and tables by the Princeton Physics Department. It seems, then, that these theoretical calculations form a safe basis for reliable interpolation and extrapolation, though we hope that they can soon be replaced by accurate directly measured values from high energy accelerators. We nevertheless think it desirable to give in detail the range-energy curve which we are using in our work and describe the method by which it is constructed, because it enters into various mass determinations of hyperons and K-mesons which we have recently carried out.

PROCEDURE

The following experimental data on the range-energy relation of protons in Ilford emulsions are available:—

(*a*) Gibson, Prowse and Rotblat measured the range of proton tracks in the energy interval 2 to 21 MeV. They used Ilford C-2 emulsions of thickness 200 μ and 400 μ. After placing their emulsion into a vacuum for 6 to 12 hours in the presence of phosphorous pentoxide, they accept the manufacturer's density of 3·94 gm./cm.^{3}

(*b*) Heinz used protons of 342·5 MeV from the Berkeley cyclotron. He also uses C-2 Ilford emulsions and measures the emulsion density before and after exposure. His value for the emulsion density is 3·81 gm./cm.^{3}

(*c*) Bradner, Smith, Barkas and Bishop use protons in the energy interval 8 MeV to 40 MeV. They do not mention the density of the C-2 emulsions (100 μ thick) used in their experiment. They kept their emulsions in vacuum for six minutes but without any dehydrating agent (They did not find any difference when these plates were kept from six minutes upto six hours in vacuum).

We have normalised the measured values of proton ranges to an emulsion density of 3·94 gm./cm.^{3} The range given by Heinz for 342·5 MeV protons for example would (in terms of gm./cm.^{2}) be somewhat larger if 4·4% of water had been removed from his emulsion. We take the "normalised" range in Heinz's experiment to be 93·6 $\pm$ 0·25 gm./cm.^{2} equal to 23·73 $\pm$ ·063 cm. of emulsion. The data of Bradner *et al.* cannot be so normalised because in their paper the density of the emulsions used by them is not mentioned.

We now calculate, using the Princeton curves, the ratio K between the square of the range in emulsion (expressed in mm.) and the product of the ranges in Aluminium and Lead (expressed in gm./cm.^{2}). For the range

in emulsion we use the data given by Gibson *et al.*, the normalised value of Heinz and the not-normalised values of Bradner *et al.* The results are shown in Plate VII, Fig. 1. It appears from this figure that:

(*a*) Bradner's values are only fairly consistent with each other and are not consistent with those of Gibson *et al.* Presumably they refer to emulsions of slightly varying density.

(*b*) The data of Gibson *et al.*, and the normalised point of Heinz indicate a value of K independent of energy and equal to $5{\cdot}075 \pm {\cdot}080$ (The fluctuations from the mean are of the same order as the errors involved in reading the Princeton curves).

It seems then, that the theoretical curves given by the Princeton Group, together with the assumption that the constant K is independent of energy, agree with experimental data and offer a rational procedure for interpolation and extrapolation. We have calculated various proton ranges in emulsion from the Princeton curves using $K = 5{\cdot}075$. They are given in Table I, and

TABLE I

Range of Protons in Nuclear Emulsions of Density 3·94 *gm./cm.*[3]

E (MeV)	R (μ)	E (MeV)	R (mm.)
1	13·80	15	1·100
1·5	25 50	20	1·815
2	39·90	30	3·615
3	74·60	40	5·960
4	118·5	50	8·750
5	172·5	60	11·85
6	235·0	80	19·80
7	303·0	100	29·70
8	378·0	150	58·15
10	554·0	200	95·40
		300	192·0
		342·5	237·5
		400	296·5
		600	552·5
		800	831·0
		1000	1135·0

the corresponding curve is shown in Plate VIII, Fig. 2. Table I and Plate VIII, Fig. 2 refer to C-2 emulsions of density 3·94 gm./cm.[3] Since the composition of C-2 emulsions is identical with that of G-5 emulsions they are applicable to G-5 emulsions as well. In addition to the range measurements available at

various energies one accurately fixed point on the curve is obtained from μ-mesons of unique energy arising from the decay of π^+-meson at rest. Since the masses of π-meson and μ-meson are known within 0·2–0·4 electron masses[7] the μ-meson energy (4·12 MeV) is known accurately. The range R_0 of the μ-meson arising from the decay of π^+-meson at rest is $574{\cdot}5 \pm 7{\cdot}7\,\mu$ for emulsions of density 3·94 gm./cm.^{3} as read from the curve in Plate VIII, Fig. 2. The error in range reflects the errors in the π- and μ-meson masses. The range energy relation can be normalised to other G-5 or C-2 emulsions of varying moisture content by measuring the mean range R of flat μ-mesons, emitted from the decay of π^+-mesons at rest, and multiplying all observed ranges by the appropriate factor R/R_0.

Accurate measurements on the variation of the density of C-2 emulsions as a function of the relative humidity has been recently carried out by A. J. Oliver.[8] From these measurements, Barkas[9] has suggested that the density of 3·915 gm./cm.^{3} sometimes ascribed to Ilford emulsions at 50% relative humidity is too high. If, therefore, the density of 3·94 gm./cm.^{3} given by Gibson, Prowse and Rotblat to their emulsions is also too high, then we have to consider the good fit we obtained for Heinz's point on our curve as accidental. Even so, if our assumption that K is constant upto very high energies is correct, our range-energy relation will still give the correct shape to the curve; but will require proper renormalisation.

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EXPLANATION OF PLATES

PLATE VII

FIG. 1. The figure shows the experimentally determined ranges in nuclear emulsion for protons of various energies as observed by different investigators. The ordinate represents the square of the range in emulsion expressed in units of the geometric mean of the corresponding ranges in aluminium and lead. $K = \frac{R^2_{(em)}}{R_{Al} \times R_{Pb}}$ (see text).

PLATE VIII

FIG. 2. The range of protons and π-mesons in nuclear emulsions is plotted against their kinetic energy (expressed in units of their respective rest energies).

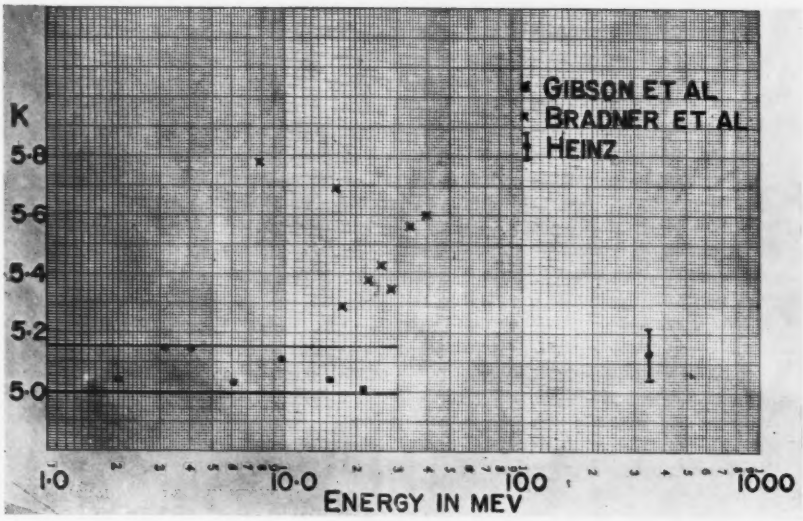
K
5·8
5·6
5·4
5·2
5·0
GIBSON ET AL.
BRADNER ET AL.
HEINZ
1·0
10·0
100
1000
ENERGY IN MEV

FIG. 1

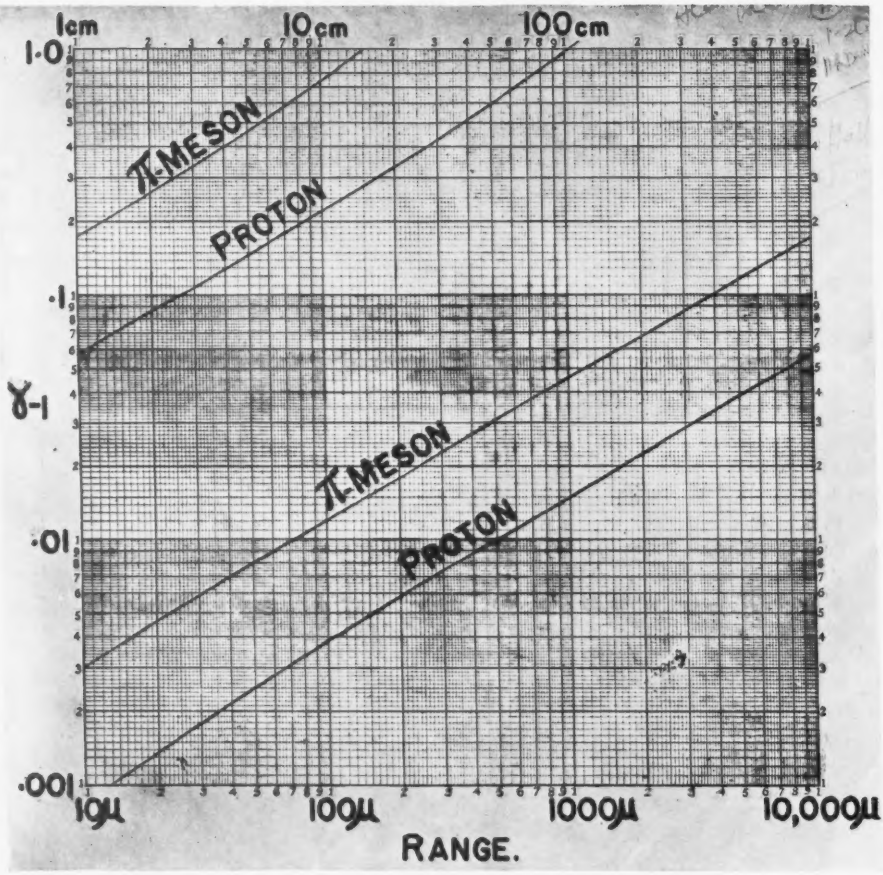
1cm
10cm
100cm
1·0
·1
·01
·001
γ-1
π-MESON
PROTON
π-MESON
PROTON
10μ
100μ
1000μ
10,000μ
RANGE.

FIG. 2

THERMAL EXPANSION OF NITRATES OF LEAD, BARIUM AND STRONTIUM

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(Communicated by Prof. R. S. Krishnan, F.A.SC.)

1. Introduction

Lead, barium and strontium nitrates form an isomorphous series of cubic crystals, which can be crystallised from aqueous solutions without water of hydration at ordinary temperatures. X-ray studies of these crystals carried out by Vegard (1922) indicate that they belong to the pentagon dodecahedral type of the tetrahedral class. The metal atoms are arranged in a face centred cubic lattice and the nitrate ions are situated on the diagonals of the elementary cubes into which the unit cell can be divided. There are four molecules per unit cell. These crystals belong to the same space group as sodium chlorate and the nitrate ions are not planar. The Raman effect in these crystals has been studied by B. L. Rao (1941), and Couture and Mathieu (1947, 1948). The stress optical coefficients for barium and lead nitrates were measured by Bhagavantam and Krishna Rao (1948, 1953). The elastic constants were determined by Bhimasenachar and Seshagiri Rao (1950).

The aim of the present work was to investigate the thermal expansion coefficients of single crystals of these nitrates and to find out the influence of the metallic ion on the coefficient of expansion.

2. Method of Measurement

The thermal expansion of these crystals above room temperature was measured by the interferometric method. The apparatus was the same as that set up by Press (1950) and used extensively by Sharma and Sridhar in this laboratory. The temperature was measured with a calibrated chromel-alumel thermocouple and a precision vernier potentiometer reading up to $\pm 1\,\mu v$. The rate of heating never exceeded 1° C. per minute. The thermo E.M.F. was read every minute and the heating current was regulated when necessary to maintain a uniform rate of heating. The time of transit of the fringe across a fixed mark was noted. The thermo E.M.F. exactly at the time a fringe crossed could be found by interpolation. Different rates of heating were also employed. The experiment was repeated after taking apart the interferometric arrangement and resetting it for a fresh run.

The crystals were grown by slow evaporation from saturated solutions of the salts. Transparent crystals of 4 to 5 mm. height were obtained. Three such pieces were ground to form pyramids of the same height and were used as the spacers between the interferometric plates. The interferometric arrangement was given a heat treatment before the commencement of each experiment.

RESULTS

A smooth curve is drawn passing evenly through the experimental points obtained in different runs and the value of the expansion coefficient $\bar{a}$ read from the curve is given in Table I. The curves are exhibited in Fig. 1.

TABLE I

Mean expansion coefficient $\bar{a}$ of barium, strontium and lead nitrates

Temperature °C.	$\bar{a} \times 10^6$		
	$Ba(NO_3)_2$	$Sr(NO_3)_2$	$Pb(NO_3)_2$
75	18·3	32·1	32·9
100	19·1	32·6	33·4
125	19·9	33·2	33·8
150	20·8	34·0	34·4
175	21·6	34·4	34·8
200	22·4	34·8	..
225	23·2	35·4	..
250	..	35·8	..
275	..	36·6	..
300	..	37·2	..
325	..	38·2	..
350	..	39·2	..
375	..	40·2	..
400	..	41·0	..
425	..	42·2	..
450	..	45·0	..
475	..	47·8	..
500	..	48·2	..
525	..	46·6	..
Lattice const. in Å.U.	8·11	7·81	7·84
Radius of Metallic ion, in Å.U.	2·17	2·04	2·05

The maximum scatter of the experimental points obtained in different runs did not amount to more than 3%. In the majority of cases the agreement was much closer.

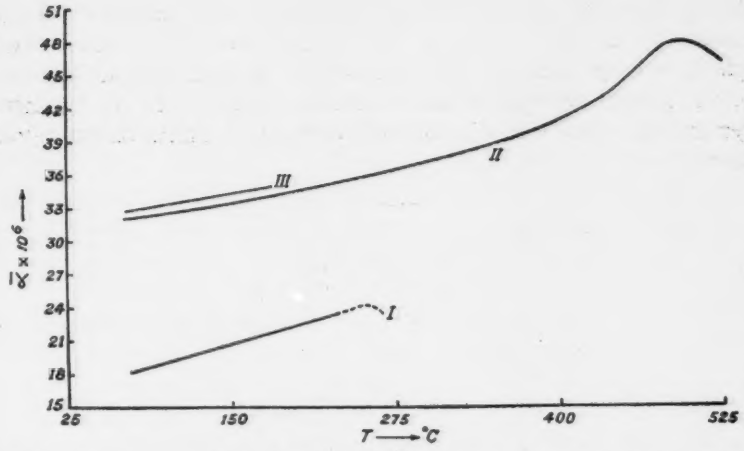

FIG. 1. Expansion Coefficient $\bar{\alpha}$ *vs.* Temperature Curve for (I) Barium Nitrate, (II) Strontium Nitrate and (III) Lead Nitrate.

Values near the room temperature were not obtained because the furnace took a certain time to reach thermal equilibrium.

The expansion coefficients are mean values over 20° C. temperature intervals.

Lead nitrate decomposes rapidly above 200° C. (Mellor, 1924). Crystals of barium nitrate cracked to pieces when they were heated beyond 270° C. But from 250° C. they gave anomalous results indicated by the dotted curve in Fig. 1. Crystals of strontium nitrate did not exhibit any such phenomena up to about 550° C.

The expansion coefficients of lead, barium and strontium nitrates could be expressed satisfactorily by the following equations:—

$Pb(NO_3)_2$: $\bar{\alpha}_t = 31{\cdot}5\times10^{-6} + 1{\cdot}9\times10^{-8}t$° C. (Between 75–180° C.)

$Ba(NO_3)_2$: $\bar{\alpha}_t = 15{\cdot}8\times10^{-6} + 3{\cdot}3\times10^{-8}t$° C. (Between 75–250° C.)

$Sr(NO_3)_2$: $\bar{\alpha}_t = 31{\cdot}6\times10^{-6} + 8{\cdot}0\times10^{-9}t$° C. $+ 4{\cdot}0\times10^{-11}t^2$ (Between 75–425° C.)

radius of oxygen atom $= 0{\cdot}75$ Å.U.

radius of nitrogen atom $= 0{\cdot}75$ Å.U.

At the bottom of Table I are entered the lattice constants and radii of the ions of these nitrates. These values are taken from Vegard (1922).

In Table II are collected the elastic constants of the crystal. The values of C_{11} and C_{12} were determined by Bhimasenachar and Seshagiri Rao (1950). The values of S_{11}, S_{12} and $S_{11} + 2S_{12}$ are calculated from these measurements. The molar volumes V are taken from Vegard (1922). The specific heat of barium nitrate was measured between 13° C. and 98° C. by Regnault (1841). The specific heats of all the three nitrates were determined by Kopp (1865) between 16° and 48° C.

According to Gruneisen (1914) the linear expansion coefficient for a cubic crystal is given by

$$V\alpha = (S_{11} + 2S_{12})\gamma C_v$$

Here C_v is the molar specific heat and γ is the Gruneisen constant. The values of γ calculated using the above formula are given.

TABLE II

Elastic constants, molar volume, molar specific heat, and Gruneisen constant of barium, strontium and lead nitrates

Substance	$C_{11}\times10^{-11}$ $\frac{\text{dynes}}{\text{sq. cm.}}$	$C_{12}\times10^{-11}$ $\frac{\text{dynes}}{\text{sq. cm.}}$	$S_{11}\times10^{13}$ $\frac{\text{cm.}^2}{\text{dyne}}$	$S_{12}\times10^{13}$ $\frac{\text{cm.}^2}{\text{dyne}}$	$(S_{11}+2S_{12})\times10^{13}$	$\bar{\alpha}\times10^{6}$ at 75° C.	Molar volume V	Molar sp. heat C_v	γ
$Ba(NO_3)_2$	5·93	1·89	20·0	− 4·8	10·4	18·3	80·9	40	0·85
$Sr(NO_3)_2$	4·73	2·18	29·8	− 9·4	11·4	32·1	72·2	38	1·28
$Pb(NO_3)_2$	4·56	3·09	48·4	−19·5	9·4	32·9	73·1	36	1·70

DISCUSSION

The expansion coefficients of strontium and lead nitrates do not differ much till 200° C.; the difference between the coefficients is small and within the limits of experimental error (3%) (see Table I).

The expansion coefficient of barium nitrate is much smaller than the coefficients for the other two isomorphous salts.

Up to the highest temperatures studied (180° for lead and 250° C. for barium nitrates) the coefficients of expansion of barium and lead nitrates increase linearly with temperature. The rate of increase is greater for barium nitrate than for the other two salts.

The expansion coefficient of strontium nitrate exhibits a parabolic increase with temperature. While up to 200° C. its rate of increase is almost the same as that of lead nitrate, at higher temperatures the rate also goes up.

Near 425° C. the expansion coefficient exhibits a sharp rise. After 500° C., the curve exhibits a fall. The melting point of strontium nitrate is at 570° C.

The strontium and lead salts possess identical lattice constants and radius of the metallic ion. Hence a substitution of strontium for lead cannot be expected to cause any great change in the forces binding the different units in the lattice. This conclusion is in agreement with the fact that these two salts possess identical expansion coefficients (see Table I).

Barium ion has a larger radius than strontium or lead and hence a substitution of barium for the other ions might be expected to loosen up the lattice. So we should expect barium nitrate to have a slightly larger expansion coefficient than the other two salts. The contrary is the case. The expansion coefficient of barium nitrate is considerably smaller than that of the other two salts. This indicates that in spite of the larger radius, the binding in barium nitrate is tighter than in the other two. The melting point of barium nitrate, 592° C., is the highest among the three salts. Also a comparison of the solubilities of the three nitrates in water is interesting. The solubilities at 30° C. of barium, strontium and lead nitrates are 11·6, 88·6 and 60·7 gm. per 100 c.c. of water. The very small solubility of barium nitrate indicates a stronger binding in the crystal. This is also supported by the following facts. The lattice lines appearing in the Raman spectrum of barium nitrate are found to be more intense than those of strontium or lead nitrates (Couture and Mathieu, 1948). A study of the magneto-optic anomaly of aqueous solutions of these salts (Ramaseshan, 1950) shows that the anomaly factor is 0·651 for barium, 0·433 for strontium and 0·503 for lead nitrates. The higher value in barium nitrate solution definitely points to a stronger binding in this salt.

In Table II the values of Gruneisen's constant have been calculated for the three crystals. We find γ for barium nitrate is much less than 1 while the γ values of strontium and lead nitrates are of the order to be expected for truly ionic crystals. Small values of γ are typical for homopolar crystals. Probably the binding in barium nitrate is not truly ionic but partly homopolar. A study of the magneto-optic anomaly in these crystals may be expected to throw some light on this point.

In conclusion the author wishes to thank Prof. R. S. Krishnan for his continued interest and encouragement throughout the course of this work.

Abstract

The expansion coefficients of barium, strontium and lead nitrates have been studied above room temperatures. Lead nitrate decomposes rapidly

beyond 200° C. while crystals of barium nitrate crack above 270° C. Strontium nitrate exhibited no such phenomena till 550° C.

These crystals are isomorphous. The expansion coefficients of lead and strontium nitrates are nearly equal; they also possess the same ionic radii. Barium nitrate has a considerably smaller expansion coefficient though its ionic radius is larger than for the other two. This peculiar behaviour of barium nitrate indicates a stronger binding in this crystal—a fact which is supported by measurements on the elastic constants, Raman effect, solubilities of these salts and a study of the magneto-optic anomaly of aqueous solutions of these salts.

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A GENERALISED THEORY OF THE CHRISTIANSEN EXPERIMENT

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1. Introduction

In a paper[1] published in these *Proceedings* nearly six years ago, the theory of the well-known Christiansen experiment was discussed on a wave-optical basis. The expression derived in that paper for the transmission coefficient of a Christiansen light filter showed it to be an exponential function involving five variables, namely the wave-length of the light, the thickness of the cell, the size of the individual particles of the powder, the difference between the refractive indices of the powder and the surrounding liquid and finally also the proportions of the volume of the cell occupied respectively by the liquid and by the particles of the powder. In the present paper, it is proposed to deal with the more general case in which the particles of the powder are *birefringent* and hence their refractive index varies with the orientation of the crystallites within the cell. The mathematical treatment adopted is on much the same lines as that followed by us in discussing the theory of the propagation of light in polycrystalline media.[2] The only difference, in fact, is that some of the cubical elements of volume each of edgelength Δ which we imagine the cell to consist of must now be considered as being filled either by the liquid of refractive index μ_l or by the crystallites. These latter are assumed to be of cubical shape and to have their edges parallel to the three optic directions for which the refractive indices are μ_1, μ_2 and μ_3 respectively. We also assume the incident light beam to be plane-polarised with its vibration direction parallel to one set of edges of the cubical blocks and that the operative refractive index of any one block may be either μ_1, μ_2 or μ_3 with equal probabilities if it is a crystallite or μ_l if it is filled with the liquid.

2. Mathematical Formulation

We shall denote by p and q the respective probabilities of a cubical block having anyone of the three principal refractive indices μ_1, μ_2, μ_3 of the crystallites and of the refractive index μ_l of the liquid. Then $3p$ and q would represent the proportion of solid and liquid elements in the Christiansen filter and therefore

$$3p + q = 1 \qquad (1)$$

As before, we consider a typical case in which the incident wave-train which we shall represent by

$$y = e^{\frac{2\pi i}{\lambda}(ct-Z)} \tag{2}$$

encounters in its passage through the Christiansen cell $(n - m)$ elementary cells of solid blocks and m cells of liquid elements. The probability of the occurrence of this event is obviously

$$\frac{n!}{(n-m)!\, m!}\ (3\,p)^{n-m}\, q^m \tag{3}$$

If further in any specification of the state of orientation of the crystallites inside the filter, k_1, k_2 and k_3 of the $(n - m)$ cubical blocks considered above have refractive indices μ_1, μ_2 abd μ_3 respectively, then the optical path retardation of the emergent wave for this configuration is $(k_1\mu_1+k_2\mu_2+k_3\mu_3)\,\Delta + m\mu_l\Delta$. We have in addition

$$(k_1 + k_2 + k_3) = (n - m) \tag{4}$$

Also, the probability of occurrence of a state in which k_1, k_2 and k_3 cells in a row of $(n - m)$ cells can be orientated so as to have refractive indices μ_1, μ_2 and μ_3 is

$$\frac{(n-m!)}{k_1!\, k_2!\, k_3!}\left(\frac{1}{3}\right)^{n-m} \tag{5}$$

Combining (3) and (5) we find that the proportion of area of the rear surface of the filter from which an emergent wave described by

$$e^{\frac{2\pi i}{\lambda}(ct-Z-\overline{k_1\mu_1+k_2\mu_2+k_3\mu_3+m\mu_l}\Delta)} \tag{6}$$

proceeds is equal to

$$\frac{n!}{k_1!\, k_2!\, k_3!\, m!}\ p^{n-m}\, q^m \tag{7}$$

The disturbance emerging from the Christiansen cell can now be obtained by a superposition of all the different wave functions of the type (6) multiplied by suitable weight factors of which (7) is a typical example. Hence,

$$y = \sum_{k_1,\,k_2,\,k_3,\,m} \frac{n!}{k_1!\, k_2!\, k_3!\, m!}\ p^{n-m}\, q^m\, exp\ \frac{2\pi i}{\lambda}(ct - Z - \overline{k_1\mu_1+k_2\mu_2+k_3\mu_3+m\mu_l}\Delta)$$

$$= e^{\frac{2\pi i}{\lambda}(ct-Z)}\left\{p\left(e^{\frac{-2\pi i}{\lambda}\mu_1\Delta} + e^{\frac{-2\pi i}{\lambda}\mu_2\Delta} + e^{\frac{-2\pi i}{\lambda}\mu_3\Delta}\right) + qe^{\frac{-2\pi i}{\lambda}\mu_l\Delta}\right\}^n \tag{8}$$

The average refractive index of the medium is now

$$\mu = p(\mu_1 + \mu_2 + \mu_3) + q\mu_l \tag{9}$$

If the birefringence of the crystalline particles is small and if further the three refractive indices of the solid powder do not differ much from the index of the liquid, then an approximation for (8) for large values of n can be effected by proceeding exactly as in the preceding paper. Denoting by d the total thickness of the cell, and neglecting terms of order higher than two in the differences between the various refractive indices, we can rewrite (8) as

$$y = e^{\frac{2\pi i}{\lambda}(ct - Z - \mu d)} \left[1 - \frac{2\pi^2\Delta^2}{\lambda^2}\left\{\sum_{r=1}^{3} p_r(\mu_r - \mu)^2 + q(\mu_l - \mu)^2\right\}\right]^n \tag{10 a}$$

Now if $\mu_1, \mu_2 \ldots \mu_n$ are n quantities having the respective probabilities of occurrence $p_1, p_2 \ldots p_n$ in any observation, then

$$\sum_{\substack{r,s \\ r<s}} p_r p_s(\mu_r - \mu_s)^2 = \tfrac{1}{2}\sum_{r,s=1}^{n} p_r p_s(\mu_r - \mu_s)^2$$

$$= \sum_s p_s\left(\sum_r p_r\mu_r^2\right) - \left(\sum_r p_r\mu_r\right)^2$$

$$= \sum_r p_r\mu_r^2 - \mu^2$$

$$= \sum_r p_r(\mu_r - \mu)^2$$

where μ is the average of the n quantities $\mu_1, \mu_2, \ldots \mu_n$.

Applying the above result to (10) we find that for large values of n

$$y = \mathrm{R}e^{\frac{2\pi i}{\lambda}(ct - Z - \mu d)} \tag{10 b}$$

where

$$\mathrm{R} = exp\ \frac{-2\pi^2\Delta d}{\lambda^2}\{p^2.\Sigma(\mu_2 - \mu_3)^2 + pq\,\Sigma(\mu_1 - \mu_l)^2\} \tag{11}$$

The ratio of the intensity of the transmitted light to that of the incident radiation is therefore given by

$$\frac{\mathrm{I}}{\mathrm{I}_0} = \mathrm{R}^2 = exp\ \frac{-4\pi^2\Delta d}{\lambda^2}\{p^2\,\Sigma(\mu_2 - \mu_3)^2 + pq\,\Sigma(\mu_1 - \mu_l)^2\} \tag{12}$$

Formula (12) expresses the extinction coefficient of light in its passage through the Christiansen cell in terms of several variables, namely, the wavelength of the light employed, the thickness of the cell, the size of the crystalline particles, the birefringence of the same and the three differences between the refractive index and the three principal indices of the birefringent material, and finally the proportion of the liquid and solid elements in the cell. By giving suitable values to p, q, μ_1, μ_2 and μ_3, several interesting cases of special importance can be deduced from (12). Thus we observe that the case of the polycrystalline aggregate considered earlier follows readily from (12) if $q = 0$ and $p = \frac{1}{3}$. Again, by writing $\mu_1 = \mu_2 = \mu_3$; $p = \frac{\sigma}{3}$ and $q = (1 - \sigma)$ which corresponds to the case of a Christiansen cell composed of isotropic particles mixed in a liquid in the proportion $\sigma : (1 - \sigma)$, we obtain the result

$$I = I_0 \, exp \frac{-4\sigma(1-\sigma)\pi^2 \Delta d}{\lambda^2} (\mu_1 - \mu_l)^2 \qquad (13)$$

derived earlier by one of us[1] on different theoretical grounds. By writing $\mu_1 = \mu_2 = \mu_3$; $p = \frac{1}{6}$ and $q = \frac{1}{2}$ in (12) or $\sigma = \frac{1}{2}$ in (13) we get the expression

$$I = I_0 \, exp \frac{-\pi^2 \Delta d}{\lambda^2} (\mu_1 - \mu_l)^2 \qquad (14)$$

for transmission by a cell composed of isotropic particles and a liquid of nearly the same refractive index mixed in equal proportions.

3. SOME FURTHER REMARKS

Considering once again the general formula (12), we may draw attention to certain features which we may expect to observe in the Christiansen experiment with birefringent powders differing from those noticeable when isotropic powders are employed. In the latter case, the transmission would be complete for the particular wave-length for which the solid and liquid have equal refractive indices and would fall off rapidly on either side of such wavelength. The light not transmitted by the cell would appear as a diffusion halo surrounding the direction of the optical image of the source as seen through the cell. Brilliant chromatic effects are accordingly to be expected and are indeed observed with isotropic powders in the experiment.

Formula (12) shows clearly that the effect of the birefriengence of the powder is to diminish the intensity of the transmitted light for all wavelengths, and we cannot therefore, expect any observable transmission through the cell with strongly birefringent powders, unless the size of the particles be very small and the thickness of the cell be reduced to a minimum. In

such cases the colour of the transmitted light would be determined predominantly by the factor $1/\lambda^2$ appearing in the argument of the exponential. Hence, it would be reddish in colour and the chromatic effects observed with isotropic powders would be absent. In these circumstances, the liquid in the cell serves only to secure optical continuity between the discrete particles contained in the cell. On the other hand, if the birefringence be small, one may expect to observe chromatic effects similar to those observed with isotropic powders. It would be necessary, however, to work with fairly fine powders and moderate cell thickness for noticeable transmission to occur. The formula also shows that the maximum transmission in these circumstances would be exhibited for those wave-lengths for which the refractive index of the liquid is most nearly equal to a species of average of the three indices of the crystal. But this is not the average index in the ordinary sense of the word.

Another important consequence of the formula is that even if the birefringence of the powder be not very small, chromatic effects would be observable when the proportion of the volume in the cell occupied by the powder is sufficiently small. In the usual form of the Christiansen experiment, the particles are allowed to settle down and form a compact aggregate at the bottom of the cell. To observe the effects now contemplated, the contents of the cell should be stirred up; alternatively the particles should be so small that they remain suspended for a long time within the liquid. In other words, dilute suspensions of strongly birefringent powders may be expected to give brilliant chromatic effects in a Christiansen cell. However, our theory could hardly be expected to give more than a qualitative indication of the phenomena then noticeable.

Finally, we come to the question of the state of polarisation of the transmitted light as also of the diffracted light when observed with birefringent powders. The present theory indicates that if the light incident on the cell be plane-polarised, both the transmitted and the diffracted light should also be perfectly plane-polarised. So far as the transmitted light is concerned, there can be no doubt that the theoretical result is correct. For, any ellipticity consequent on the passage of light through an arbitrarily orientated crystallite would give rise to a component perpendicular to the original vibration direction in the diffracted light. But such components cannot appear in the light transmitted by the cell in the true optical sense. Indeed, provided the birefringence is small and the size of the particles and the thickness of the cell are moderate we may expect also to find that the diffraction halo is itself strongly polarised. In other circumstances, however, especially

when the particles are strongly birefringent, the diffracted light would exhibit a marked imperfection of polarisation by reason of the ellipticity effects which have dropped out of consideration in the present treatment of the problem.

SUMMARY

A formula is derived for the transmission coefficient of a Christiansen cell containing particles of a birefringent material whose interstices are filled up by a liquid of suitably adjusted refractive index. The consequences of the formula and especially the influence of the birefringence on the spectral character of the transmitted light are discussed.

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THE CHRISTIANSEN EXPERIMENT WITH BIREFRINGENT POWDERS

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1. Introduction

In the well-known experiment due to Christiansen which has been applied in the construction of monochromatic light-filters, an isotropic transparent solid, *e.g.*, optical glass, is powdered and placed inside a flat-sided glass cell and the latter is then filled up by a liquid whose refractive index is adjusted to equality with that of the solid for some particular wave-length in the spectrum. Christiansen himself tried using powdered quartz in the experiment, and found that it did not prove a success. The light entered the cell and was diffused by the powder-liquid mixture, but the source of light could not itself be seen through the mixture. Considered from the standpoint of geometrical optics, this result is not surprising, since the refractive index of the particles of a birefringent powder would depend on their orientation within the cell and hence would vary from particle to particle.

However, geometrical optics does not correctly describe what is actually observed in the Christiansen experiment even with optically isotropic powders,[1, 2] and hence there is no reason to believe that it would be any more successful in the case of birefringent powders. In another paper[3] appearing in the present issue of these *Proceedings*, a general theory has been developed which indicates that chromatic effects analogous to those observed with isotropic powders should also be capable of appearing with birefringent powders in appropriate circumstances. The manner in which the birefringence would modify the observed effects has also been discussed in that paper.

It is proposed in what follows to place on record some observations which we have made broadly confirming the indications of the general theory. It has been found that provided the birefringence is fairly small and the material is in a finely subdivided state, it is possible to obtain a true transmission exhibiting brilliant colours. It is also found that the light so transmitted is not greatly inferior in the degree of its monochromatism to that observed with isotropic material in similar circumstances. Another noteworthy feature is that if the light incident on the cell is plane-polarised, the

light transmitted by it is also completely plane-polarised. This might seem paradoxical when it is recalled that the light has had to pass through considerable thicknesses of a birefringent material; nevertheless the observation is in strict accord with the theory. On the other hand, the diffusion halo appearing in directions surrounding the regularly transmitted light exhibits imperfect polarisation to an extent depending on the fineness of the subdivision of the material. The colours exhibited by the diffusion halo are also found to be markedly different for the components of the light vibration respectively parallel and perpendicular to that in the incident light.

2. SOME GENERAL OBSERVATIONS

Besides the factors which determine the transmission coefficient with isotropic powders, an additional factor appears in the present case, namely the magnitude of the birefringence of the material. The importance of this relatively to the differences in the refractive indices of the solid and liquid varies with the proportion of the volumes occupied respectively by the powder and the liquid in the cell. By varying this proportion, we may pass from one extreme case of a polycrystalline aggregate in which the liquid serves merely to secure optical continuity between the particles of the powder to the other extreme case in which the liquid occupies the whole volume except for the particles of solid held in suspension. The most interesting cases are however those in which the two components are present in nearly the same proportions. To observe chromatic effects in such cases, it is necessary to use material which is not too highly birefringent and which is in a fine state of subdivision. The latter condition is most conveniently secured by using a substance which is commercially obtainable in the state of a fine powder and hence does not need any further preparation. We shall content ourselves here by mentioning three such substances which we have found to work very well in the experiment.

Barium sulphate can be used with success in a cell from five to ten millimetres thick with carbon disulphide as the liquid filling it up. The addition of a few drops of benzene shifts the transmission from the yellow towards the violet end of the spectrum. Precipitated calcium sulphate in the form of gypsum also gives good results, the appropriate liquid in this case being monochlorobenzene; a drop or two of carbon disulphide shifts the transmission towards the red, while the addition of a few drops of benzene shifts it towards the violet. Magnesium fluoride also serves admirably; the appropriate liquid to use in this case is acetone, the addition to which of benzene causes the transmission to appear first at the violet end of the spectrum and shifts it step by step toward the red.

3. Polarisation Phenomena

In all the three cases mentioned, beautiful chromatic haloes are observed surrounding the direction of the transmitted light. The colour of the halo varies with the direction of observation and also alters when the spectral region of transmission is shifted. Viewing a bright and well-defined light source through the cell held before the eye of the observer, with one polaroid inserted between the source and the cell and a second polaroid between the cell and the observer's eye, striking polarisation phenomena may be observed. In all cases, the image of the light source is itself completely extinguished when the two polaroids are crossed. But the diffusion halo seen surrounding the light-source shows imperfect polarisation. The magnitude of this imperfection differs very much in the case of the three substances mentioned above. With magnesium fluoride the halo disappears almost completely when the polaroids are crossed. With barium sulphate its extinction is less complete, and the diffusion halo remains observable in directions adjacent to the source and gives indications of a bright cross with its arms bisecting the angle between the vibration directions of polariser and analyser. The extinction of the diffused light is least perfect in the case of calcium sulphate and the halo remains observable over the whole area of the field even with the polaroids crossed.

4. Observations with Powdered Quartz

The following method was adopted to prepare quartz in a state of fine subdivision but uncontaminated by extraneous material. A transparent piece of crystalline quartz was heated and dropped into cold distilled water. The fragments into which it broke as the result of this treatment were heated in a silica dish and then again dropped into cold distilled water. Repetition of this procedure reduced the substance to a state of powder and the material thus obtained was then ground up very fine between two quartz crystals with flat faces and finally separated into four grades by elutriation in distilled water. The two finest grades thus obtained were those which remained in suspension in a tall beaker of distilled water for ten and twenty minutes respectively.

The optical effects exhibited by the four grades of quartz when placed in a cell and filled up with a mixture of benzyl alcohol and carbon disulphide were found to be very different. The first or roughest grade gave only a diffusion halo without any regular transmission even with a cell only one millimetre thick. With the second grade of powder and the same thickness of cell, a very weak transmission can be glimpsed, the bright diffusion halo overlying it making the observation rather difficult. On the other hand,

the two finer grades when allowed to settle down in a cell two millimetres thick show brilliantly coloured transmitted images of the source. With a cell only one millimetre thick, the transmission is even more brilliant, but its colour is then less saturated.

In all cases, if a transmission is obtained at all, it is completely extinguished when the cell is placed between two crossed polaroids. On the other hand, the appearance of the diffusion halo as well as its state of polarisation shows remarkable variations with the grade of powder and the thickness of cell employed. With the coarser grades of material, the halo is found to be completely depolarised. On the other hand, with the finer grades the halo shows a very marked degree of polarisation. Not merely the brightness of the halo but also the distribution of colour in it is strikingly different when the polaroids are respectively parallel and crossed. This difference is best described by the statement that when the polaroids are crossed, the halo exhibits colours similar to those of the transmitted light (which is extinguished in the same circumstances); *per contra*, with the polaroids parallel, the colour of the halo is complementary to that of the transmitted light. These changes were most striking when observed with the cells of smaller thickness. For, with such cells, the colour of the halo over its whole area is markedly different from that of the transmitted light and exhibits its complementary character most clearly. The changes produced by the rotation of the analyser are therefore particularly striking.

5. OBSERVATIONS WITH DILUTE SUSPENSIONS

With the two finer grades of quartz powder, it is possible to use much thicker cells with success for observing the transmitted light, if they are filled with an excess of liquid in which the powder is held as a dilute suspension. By varying the thickness of the cell and the quantity of material held suspended in it, one can either increase or decrease the saturation of the colours observed in the transmitted light and in the diffusion halo. It is worthy of note that in such cases the diffusion halo exhibits a colour complementary to that of the transmitted light even in directions adjacent to the latter. This indeed is what optical theory indicates should be the case for a dilute suspension. On the other hand, multiple scattering comes into play in the case of dense aggregates, and the colour of the halo is in consequence almost indistinguishable from that of the transmitted light in closely adjoining directions and only further out changes by insensible gradations to the complementary tint.

It appears worthwhile also to record some observations made with strongly birefringent powders, calcium carbonate in the form of precipitated

chalk being a typical example. It is not possible to obtain any transmission with this material in the usual form of the Christiansen experiment. But interesting effects may be observed even with cells of considerable thickness if they are filled with carbon disulphide into which a little precipitated chalk is put in and stirred so that the liquid appears as a milky-white suspension. A bright source of light can be seen through such a suspension and exhibits a deep red colour, as indeed it should according to the theory. Here again the transmitted light is completely extinguished if the cell is placed between crossed polaroids. On the other hand, the light diffused by the cell is depolarised, but if the suspension is very dilute, the cell exhibits a bright cross between crossed polaroids. If benzene is added to the suspension of chalk in carbon disulphide, thereby bringing the index of the liquid mixture nearer to the mean index of calcium carbonate, the colour of the transmitted light is shifted towards shorter wave-lengths in the spectrum.

6. Description of the Figures in Plate IX

By way of illustration of the foregoing observations, some photographs have been reproduced in Plate IX accompanying this paper. The following remarks are explanatory notes on the same.

Fig. 1 shows the appearance of a small brilliant light-source as viewed through a cell five millimetres thick containing barium sulphate in a finely subdivided state. This had settled down at the bottom of a mixture of carbon disulphide and benzene so adjusted as to transmit the green part of the spectrum. The picture clearly shows the transmitted light and the diffusion halo surrounding it.

Fig. 2 shows the same experiment with the cell placed between crossed polaroids and photographed with a much longer exposure. The source itself is extinguished in these circumstances. The halo also disappears except at its brightest part near the centre which exhibits the bright cross whose arms bisect the angle between the vibration directions of the polariser and analyser.

Fig. 3 reproduces the spectra of the light transmitted by the cell containing barium sulphate in the same circumstances. The colour of the transmitted light was shifted in steps from orange-yellow to blue by the successive additions of benzene to carbon disulphide. The lines of the mercury arc spectrum are reproduced to indicate the positions of the transmission band.

Fig. 4 is a photograph of a small bright source of light seen through a cell two millimetres thick containing the finest grade of quartz powder

which had settled down at the bottom of the cell. Benzyl alcohol to which a few drops of carbon disulphide had been added was the liquid used and the mixture transmitted the yellowish-green part of the spectrum. The photograph itself was rather overexposed with the result that the transmission and the halo are not seen clearly distinguished from one another. The streaky nature of the halo is clearly shown.

Fig. 5 is a photograph of a bright source of light seen through a thin film of nitrobenzene containing fine particles of lithium carbonate in suspension and held between crossed polaroids. The dark arms of the cross seen in the diffusion-halo are parallel to the vibration directions of the polariser and analyser respectively.

7. SUMMARY

Optical effects analogous to those exhibited by isotropic materials in a Christiansen cell are also observable with birefringent materials in a fine state of subdivision. While the transmitted light is fully polarised, the diffusion halo is depolarised in part and exhibits colours between parallel and crossed polaroids which are complementary to each other.

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FIG. 1

FIG. 2

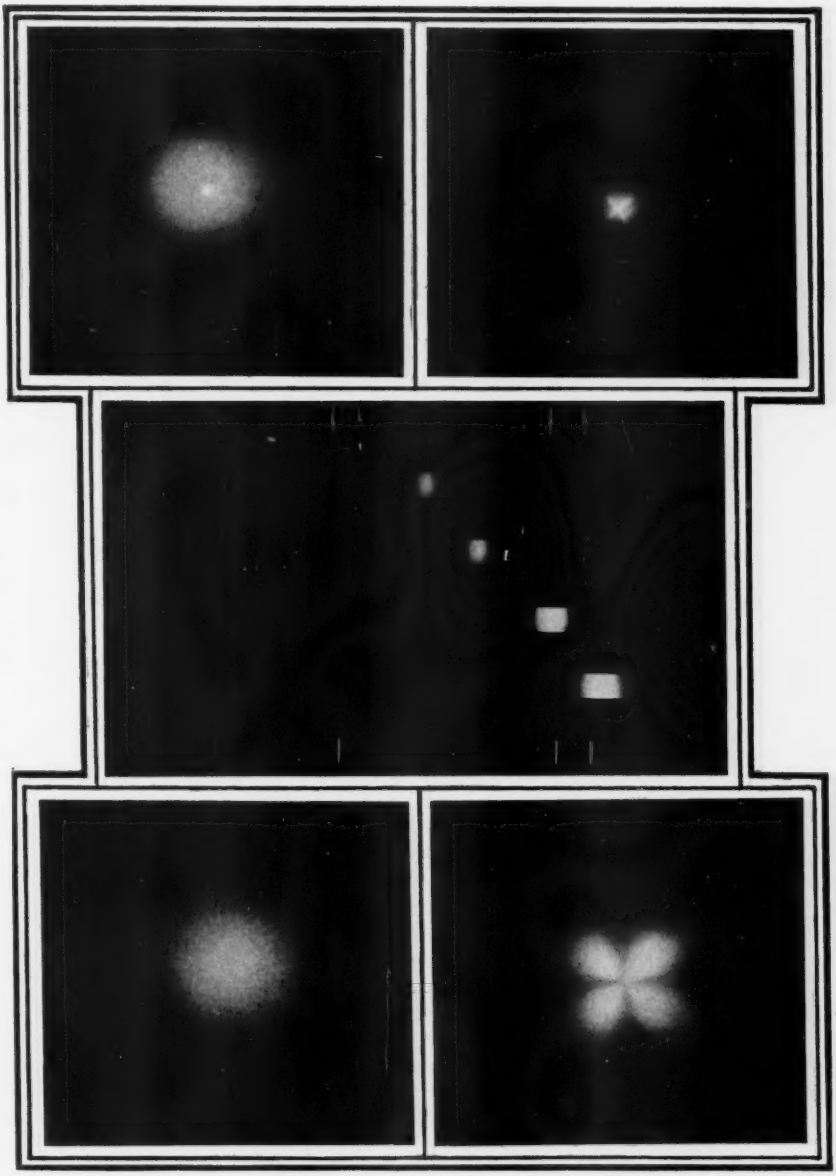

FIG. 3

FIG. 4

FIG. 5

Christiansen Experiment with Birefringent Powders.

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